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N. S. AKULOV'S THEORY OF CHAIN PROCESSES

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[The following report is a summary of a USSR review of the book "Teoriya Tsepnykh Protsessov" (Theory of Chain Processes) by N. S. Akulov, State Publishing House of Technical and Theoretical Literature, Moscow, 1951.]

Notwithstanding the great importance of the theory of chain processes, only a very limited number of monographs and manuals on this subject is available in world literature. Akulov's monograph, which represents a result of 10 years of work by Akulov and his school, fills out this blank to a considerable extent. Although the work done by N. A. Shilov, D. V. Alekseyev, Bodenstein, and N. N. Semenov has advanced the theory of chain processes to a considerable extent, this theory until recently still remained on a level where chain processes in the main had to be studied qualitatively rather than quantitatively. In chain processes which actually take place, particularly chain processes that are involved in reactions that occur within gas mixtures, the intermediate active centers approach the walls of the vessel as a result of diffusion and are either adsorbed there or enter into reaction with those molecules of the initial substances that have already been adsorbed. If the intermediately formed particles disappear in reactions that occur on the walls, their concentration in the vicinity of the walls will be lower than in the center of the vessel. As a result there will be a flow of the particles from the center to the walls. In diffusing, the particles undergo mutual transformations, i.e., there is a transmutation effect.

Prior to Akulov's work, equations of the Shilov-Bodenstein type were used in interpreting chain processes. Since these equations disregard diffusion, they were not very well suited for the formulation of a theory that would explain the combustion of gas mixtures. The same [limitation] applies to Sorokin's equations.

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Although Sorokin's equations take into consideration diffusion and the multiplication of particles of a single kind, they cannot be used for the calculation of chain transformations.

The system of equations proposed by Akulov in 1947, which is free of these defects, forms the basis of the theory outlined in the second part of his monograph. Although the discussion of the mathematical methods used in the solution of equations of chain diffusion will be of interest primarily to physicists and mathematicians, the final results are simple enough. They can be applied extensively by physical chemists, engineers, and biochemists. In the second part of the monograph the author also discusses the method of cycles developed by him. This method is distinguished both by its simplicity and clarity. For the first time in the literature of chemical kinetics, modern mathematical methods have been applied by Akulov. In connection with a number of problems, for example the diffusion of particles having different coefficients of diffusion and the dependence of the rate of propagation of chain explosions on the diameter of the channel, Akulov has developed new mathematical methods.

Very interesting and important is the connection that has been established in the monograph between A. M. Lyapunov's criteria of stability and the explosion criteria. The problems formulated in connection with this approach may lead to new and important results in the field of nonlinear chain processes. The most important results were achieved by applying the equations of Akulov's chain theory to problems of combustion. Prior to Akulov's work, reliance was placed mainly on semiempirical relationships as far as pressure limits for the propagation of explosions and pressure limits for explosions in closed vessels are concerned. These relationships were formulated by D. V. Alekseyev for the propagation of explosions and by N. N. Semenov for explosions in closed vessels. A serious fault of Semenov's semiempirical equations is that their validity is restricted to low pressures only, while in practice equations which can be applied in the high pressure range are required. The problem of finding general equations which are valid both for high pressures and low pressures has been solved by Akulov. The questions involved here are discussed in detail in the text of Akulov's book.

In the fourth part of the book, the role of chain processes in various forms of the movement of matter, the evolution of precellular forms of life, and biological phenomena of various types is discussed. The author arrives at important conclusions which are in accordance with the tenets of dialectical materialism.

In addition to the positive aspects mentioned, the book also has some drawbacks. Some of the faults of the book are that the exposition given by the author bears the character of an outline and that the fourth part of the book, which deals with biological problems, does not cite enough factual data. In order to explain the existence of three limits, the author uses the simplest schemes of chain processes. Although these schemes explain the existence of three limits, very rigid conditions must be observed. Actually one may name a number of much more general chain processes, which proceed over radicals and atoms, and furnish an explanation of the three limits without observation of the rigid conditions mentioned. This has been demonstrated by Akulov in his subsequent work (Doklady Akademii Nauk SSSR Vol 83, 3, 1952).

Although Akulov in his book possibly overestimates the role which chain processes play in nature, he has formulated with considerable audacity the problems of chain processes, has given a thorough mathematical treatment to the theory of these processes, and was the first to arrive at a logically consistent solution of the problems involved. Akulov's theory led to a whole series of new relationships which are in quantitative agreement with a huge amount of experimental material accumulated by a number of investigators. It is of especial importance that this theory has furnished for the first time a correct explanation of various phenomena and of their interrelationship. This applies to the limits of spontaneous ignition, the limits of the propagation of explosions, and the limits of heat explosions.

Akulov deserves credit for establishing the priority of native science in the discovery of chain reactions. He has pointed out the work done by N. A. Shilov, D. V. Alekseyev, and other Soviet scientists and the role it has played in the formulation and development of the theory of chain reactions.

Akulov's monograph was the first to give a serious blow to P. Duhem's idealistic theories, which have shown a tendency to penetrate into physical chemistry. Even now attempts are being made to smuggle in Duhem's ideas in connection with the problem of false equilibria: it is asserted that meta-stable states of a system may exist during which chemical processes are stopped. In his treatment of the autogenesis of explosion processes, Akulov demonstrated that equilibria which may be described as false do not occur in nature.

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